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From the

INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

11 FEB 2005

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To:

BURFORD, Anthony F.
W. H. Beck, Greener & Co.
7 Stone Buildings
Lincoln's Inn
London WC2A 3SZ
GRANDE BRETAGNE

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NOTIFICATION OF TRANSMITTAL OF
THE INTERNATIONAL PRELIMINARY
EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing
(day/month/year)

09.12.2004

Applicant's or agent's file reference
AFB/JAS/P9096WO

IMPORTANT NOTIFICATION

International application No.

PCT/GB-03/03403

International filing date (day/month/year)

06.08.2003

Priority date (day/month/year)

13.08.2002

Applicant

MID-AMERICA COMMERCIALIZATION CORP. et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability requirements for enabling disclosure, clarity and support for the claims.

ALECTO

DEC 28 2004

ENTERED BY

Name and mailing address of the international
preliminary examining authority:



European Patent Office
D-80298 Munich
Tel. +49 89 2399 - 0 Tx: 523656 epmu d
Fax: +49 89 2399 - 4465

Authorized Officer

Almalé Murillo, J-A

Tel. +49 89 2399-8059



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INTERNATIONAL PRELIMINARY EXAMINATION REPORT
(PCT Article 36 and Rule 70)



Applicant's or agent's file reference AFB/JAS/P9096WO		FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/PEA/416)
International application No. PCT/GB 03/03403	International filing date (day/month/year) 06.08.2003	Priority date (day/month/year) 13.08.2002
International Patent Classification (IPC) or both national classification and IPC C10G2/00		
Applicant MID-AMERICA COMMERCIALIZATION CORP. et al.		

- This International preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
- This REPORT consists of a total of 6 sheets, including this cover sheet.

☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

 These annexes consist of a total of 6 sheets.

- This report contains indications relating to the following items:
 - I ☒ Basis of the opinion
 - II ☐ Priority
 - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
 - IV ☐ Lack of unity of invention
 - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
 - VI ☐ Certain documents cited
 - VII ☐ Certain defects in the international application
 - VIII ☐ Certain observations on the international application

Date of submission of the demand 12.03.2004	Date of completion of this report 09.12.2004
Name and mailing address of the International preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Rumbo, A Telephone No. +49 89 2399-8407 

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB 03/03403

I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, Pages

1-30 as originally filed

Claims, Numbers

1-29 filed with telefax on 07.09.2004

Drawings, Sheets

1/3-3/3 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

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5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	1-18
	No: Claims	19-29
Inventive step (IS)	Yes: Claims	
	No: Claims	1-29
Industrial applicability (IA)	Yes: Claims	1-29
	No: Claims	

2. Citations and explanations

see separate sheet

SECTION V

1. STATE OF THE ART AND NOVELTY OF THE CLAIMED SUBJECT-MATTER

1.1 The claimed apparatus of claim 19 comprises:

- (a) a syngas generation system (for reacting methane and/or oxygen containing gas)
- (b) a syngas conversion system (for converting syngas into HC or oxygenates)
- (c) a cryogenic separation system (separating H₂ from cryogenic liquid comprising CO)
- (d) a cryogenic distillation column (for separating CO from cryogenic liquid)

All the elements of the claim (a to d above) are known and (see, e.g. D1=US-A-4 782 097 and documents cited at page 2, lines 9-14 of the description of the present application as far as all the elements of the cryogenic distillation column are concerned), provided that the intended use of the apparatus either does not add a technical feature to the claimed subject matter or does not establish any connection between them, the novelty of the independent apparatus claim 19 and the rest of the claims 20-29 (containing well known isolated elements with known technical features as methane wash columns of claims 20 and 26, hydrogenation system of claim 21, conduit means of claim 22 to 24, membrane separation systems of claim 25, hydrogen recycle of claim 27 and reactors of claims 28-29) are all known from either D1 or the rest of the documents of the prior art cited at page 2, lines 9-14 of the description of the present application and cannot be considered novel.

Claims 19-29 do not meet therefore the requirements of Article 33(2) PCT.

1.2 The wording of independent claim 1 comprises:

- (a) a process for generating syngas (comprises CO and H₂) from methane and either steam and/or an oxidant gas comprising oxygen,
- (b) a conversion process for producing higher molecular weight hydrocarbons or oxygenates,
- (c) removing offgas comprising unreacted hydrogen and unreacted CO from said syngas conversion process (b).
- (d) separating unreacted hydrogen from the offgas of (c) above via a cryogenic separation process to produce hydrogen product substantially free from unreacted CO and a first cryogenic liquid comprising unreacted CO.
- (e) the first cryogenic liquid comprising unreacted CO is treated in a cryogenic distillation column to produce separated CO and a liquid which is substantially CO-free.

1.3 The novelty of the claimed subject-matter (claim 1) has been made novel in view of D1=US-A-4 782 097, due to the introduction of the terms of claim 5 within the wording of originally filed claim 1, i.e. due to the presence of **two step cryogenic separations one of which contains a distillation column** instead of the more general description of

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cryogenic step according to the chemical Engineering Progress February 1980 pages 72-79 and October 1984 pages 53-56.

Claims 1-18 meet the requirements of novelty of Article 33(2) PCT in view of D1.

1.4 The claimed subject-matter has also been made novel in view of the rest of documents of the prior art cited in the search report. In particular D2= US-A-5 173 513 disclosing a process for the obtention of methanol from syngas in which both a hydrogen rich stream (see 94.19% v/v at table 2) and a CO rich stream (see 33.55%v/v CO at table 2) are recycled to be mixed at points 114 and respectively 115 to the gas obtained from the syngas generator. **The claimed process differs** from that of D2 due to the fact that **it uses a combination of a non defined cryogenic process giving pure Hydrogen (purity unknown) and a cryogenic liquid (composition unknown) which is again separated in a cryogenic distillation column into carbon monoxide (purity unknown) and carbon monoxide free cryogenic liquid** instead of the apparatus 152 cryogenic separation (see col.12, line 21) and dry purifier absorption tower 144).

Claims 1-18 meet the requirements of novelty of Article 33(2) PCT in view of D2.

2. The claimed subject-matter does not meet the inventive step requirements of Article 33(3) EPC.

2.1 D1 is not relevant concerning the inventive step of the claimed subject-matter insofar as no CO recycling is disclosed therein. On the contrary purge gas can be used as a residual gas (see residual gas of fig I. and item 17 of figures 4. and 5.) or as a purge gas (item 15 of figure 3)

2.2 On the contrary, D2 is of the utmost relevance as far as the inventive step of the claimed subject-matter is concerned.

In fact, the claimed subject-matter cannot be considered inventive insofar as separating CO rich stream from H₂-rich stream is known either via the process of D2 or via a cryogenic process including a cryogenic column of D3=US-A-4 217 759 (see items 24, 124, 224, 324 of figures 1 to 4, table A, claims 1 to 10) cited at page 2, lines 9-14 of the description of the present application.

Merely using a separation of a stream of CO of unknown purity from a stream of H₂ of unknown purity cannot constitute the basis of any inventive step in view of either D2 taken alone or even in view of its combination with D3.

D2 taken alone explicitly solves a problem of adjusting the CO and H₂ contents of the feedstock to the syngas conversion unit by recirculating a CO rich stream and a H₂ rich

stream.

On the contrary, the claimed subject-matter merely separates the stream of CO (of unknown composition or purity) from that of H₂ (of unknown purity) without undertaking anything therewith. Of course such cryogenic separations are well known from D3 and cannot constitute per se the basis of any kind of inventive step argument.

2.3 In the absence of the evidence that a technical problem can be solved as a consequence of the presence of a differentiating technical feature within the wording of the independent claim in a non obvious way which could not have been solved in an obvious way by the technical features of D2 or the rest of documents of the prior art (e.g. D3) the inventive step of the claimed subject-matter cannot be acknowledged. In fact modifications which do not solve any technical problem can be considered obvious modifications of the prior art. This is the case of the presently claimed subject-matter which independent claim 1 does not solve any technical problem in view of D2 or D2+D3 and does not meet therefore the requirements of Article 33(3) PCT.

CLAIMS

1. A process for producing higher molecular weight hydrocarbon compounds or oxygenates from a gas comprising methane, said process comprising:

5 generating synthesis gas ("syngas") comprising carbon monoxide and hydrogen by reaction of a gas comprising methane with steam and/or an oxidant gas comprising oxygen;

producing higher molecular weight hydrocarbon compounds or oxygenates in a syngas conversion process;

10 removing offgas comprising unreacted hydrogen and unreacted carbon monoxide from said syngas conversion process; and

separating unreacted hydrogen from said offgas or from a gas derived therefrom to produce separated hydrogen product, wherein unreacted hydrogen is separated in a cryogenic separation process to produce separated hydrogen
15 product that is substantially free of unreacted carbon monoxide and a first cryogenic liquid comprising unreacted carbon monoxide, characterised in that the process further comprises:

separating unreacted carbon monoxide from said first cryogenic liquid or from a cryogenic liquid derived therefrom in a cryogenic distillation column to
20 produce separated carbon monoxide product and substantially carbon monoxide-free cryogenic liquid.

2. A process as claimed in Claim 1 wherein the cryogenic separation process is a liquid methane wash.

25 3. A process as claimed in Claim 1 or Claim 2 wherein higher molecular weight hydrocarbon compounds are produced, said process further comprising using separated hydrogen product for hydrogenation of a fraction of said higher molecular weight hydrocarbon compounds to produce liquid hydrocarbon
30 compounds.

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4. A process as claimed in Claim 3 wherein said separated hydrogen product is used in the hydrogenation of said hydrocarbon fraction without purification.

5. A process as claimed in any of Claims 1 to 4 further comprising recycling separated carbon monoxide product for conversion into higher molecular weight hydrocarbon compounds or oxygenates.

6. A process as claimed in any of Claims 1 to 5 wherein said first cryogenic liquid further comprises argon, said process further comprising removing at least one argon-enriched stream from a location in the region of high argon concentration in the cryogenic distillation column.

7. A process as claimed in any of Claims 1 to 6 wherein the substantially carbon monoxide-free cryogen liquid is substantially pure liquid methane, said process further comprising recycling a vaporised portion of said substantially pure liquid methane for conversion into syngas.

8. A process as claimed in any of Claims 1 to 7 wherein said offgas further comprises helium, said process further comprising separating helium from said separated hydrogen product using a membrane separation system.

9. A process as claimed in any of Claims 1 to 8 wherein said offgas further comprises low molecular weight hydrocarbon compounds, said process further comprising:
cooling said offgas or a gas derived therefrom to condense said low molecular weight hydrocarbon compounds to produce liquefied petroleum gas ("LPG") and removing said LPG to produce substantially LPG-free offgas;
cooling and at least partially condensing said substantially LPG-free offgas to produce partially condensed substantially LPG-free offgas; and

separating unreacted hydrogen from a vapour portion of said partially condensed substantially LPG-free offgas in said cryogenic separation process to produce said separated hydrogen product and said first cryogenic liquid.

5 10. A process as claimed in Claim 9 wherein unreacted hydrogen is present in said first cryogenic liquid, said process further comprising separating unreacted hydrogen from said first cryogenic liquid or a cryogenic liquid derived therefrom in a further cryogenic separation process to produce separated hydrogen fuel by-product and a second cryogenic liquid comprising unreacted carbon monoxide.

10 11. A process as claimed in Claim 10 wherein the further cryogenic separation process is a liquid methane wash.

15 12. A process as claimed in Claim 10 or Claim 11 wherein said offgas further comprises helium and wherein separated hydrogen product is recycled for conversion into higher molecular weight hydrocarbon compounds, said process further comprising removing a portion of said separated hydrogen product once the concentration of the helium in the separated hydrogen product is between from 1 mol % to 20 mol %.

20 13. A process as claimed in Claim 12 wherein said portion is adjusted in flowrate so that the helium concentration reaches the required concentration.

25 14. A process as claimed in any of Claims 1 to 13 further comprising at least partially vaporising liquid nitrogen ("LIN") to provide refrigeration duty to keep the process in heat balance.

30 15. A process as claimed in any of Claims 1 to 14 wherein the oxidant gas is oxygen with a purity below 99 mol % produced in an integrated cryogenic air separation process, said LIN being produced in said air separation process.

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16. A process as claimed in any of Claims 1 to 15 wherein separated hydrogen product is recycled for conversion into higher molecular weight hydrocarbon compounds.

17. A process as claimed in any of Claims 1 to 16 wherein said syngas is generated by partially oxidizing natural gas with oxygen and by reforming natural gas with steam.

18. A process as claimed in any of Claims 1 to 17 wherein said syngas conversion process is a Fischer-Tropsch ("FT") process.

19. Apparatus for the production of higher molecular weight hydrocarbon compounds or oxygenates from a gas comprising methane according to the process as defined by Claim 1, said apparatus comprising:

a syngas generation system for generating syngas comprising carbon monoxide and hydrogen by reaction of a gas comprising methane with steam and/or an oxidant gas comprising oxygen;

a syngas conversion system for converting syngas into higher molecular weight hydrocarbon compounds or oxygenates and producing offgas comprising unreacted hydrogen and unreacted carbon monoxide; and

a cryogen separation system for separating unreacted hydrogen from said offgas or a gas derived therefrom to produce separated hydrogen product that is substantially free of unreacted carbon monoxide and a first cryogenic liquid comprising unreacted carbon monoxide,

characterised in that the apparatus further comprises:

a cryogenic distillation column for separating unreacted carbon monoxide from said first cryogenic liquid or from a cryogenic liquid derived therefrom to produce separated carbon monoxide product and substantially carbon monoxide-free cryogenic liquid.

20. Apparatus as claimed in Claim 19 wherein the cryogenic separation system is a first liquid methane wash column.

21. Apparatus as claimed in Claim 19 or Claim 20 producing higher molecular weight hydrocarbon compounds, said apparatus further comprising a hydrogenation system for hydrogenating a fraction of said higher molecular weight hydrocarbon compounds using separated hydrogen product to produce liquid hydrocarbon compounds.

22. Apparatus as claimed in any of Claims 19 to 21 further comprising conduit means for feeding separated carbon monoxide product from said cryogenic distillation column to said syngas conversion system.

23. Apparatus as claimed in any of Claims 19 to 22 further comprising conduit means for removing an argon-enriched stream from a location in the region of high argon concentration in said cryogenic distillation column.

24. Apparatus as claimed in any of Claims 19 to 23 wherein said substantially carbon monoxide-free cryogenic liquid is substantially pure liquid methane, said apparatus further comprising conduit means for feeding substantially pure liquid methane from said cryogenic distillation column to said syngas generation system.

25. Apparatus as claimed in any of Claims 19 to 24 wherein the offgas further comprises helium, said apparatus further comprising a membrane separation system for removing helium from separated hydrogen product.

26. Apparatus as claimed in any of Claims 19 to 25 wherein unreacted hydrogen is present in said first cryogenic liquid, said apparatus further comprising a second liquid methane wash column in which unreacted hydrogen is separated from said first cryogenic liquid or a cryogenic liquid derived therefrom to produce

separated hydrogen fuel by-product and a second cryogenic liquid comprising unreacted carbon monoxide.

27. Apparatus as claimed in Claim 26 wherein said offgas further comprises helium and wherein separated hydrogen product is recycled for conversion into higher molecular weight hydrocarbon compounds or oxygenates, said apparatus further comprising conduit means for removing a portion of said separated hydrogen product having a concentration of helium between from 1 mol % to 20 mo %.

28. Apparatus as claimed in any of Claims 19 to 27 wherein the syngas generation system comprises a partial oxidation ("POX") reactor and an enhanced heat transfer reformer ("EHTR").

29. Apparatus as claimed in any one Claims 19 to 28 wherein the syngas conversion system comprises at least one FT reactor.

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